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(12) <b>PATENT</b>	(11) Application No. <b>AU 199950336 B2</b>
(19) <b>AUSTRALIAN PATENT OFFICE</b>	(10) Patent No. <b>765514</b>

(54) Title	Coloration with pearly luster pigments in the food and pharmaceutical sectors		
(51) <sup>7</sup> International Patent Classification(s)	A23L 001/275 C09C 001/24 A61K 047/02 C09C 001/36		
(21) Application No:	199950336	(22) Application Date:	1999.07.08
(87) WIPO No:	WO00/03609		
(30) Priority Data			
(31) Number	(32) Date	(33) Country	
19831869	1998.07.16	DE	
(43) Publication Date :	2000.02.07		
(43) Publication Journal Date :	2000.04.20		
(44) Accepted Journal Date :	2003.09.18		
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(56) Related Art	DE 4215367 WO 97/43346		

<p>(51) International patent classification<sup>7</sup>:</p> <p>A23L 1/275, A61K 47/02, C09C 1/24, 1/36</p>	<p>A1</p>	<p>(11) International publication number: WO 00/03609</p> <p>(43) International publication date: 27 January 2000 (27.01.00)</p>
<p>(21) International application number: PCT/EP99/04792</p> <p>(22) International filing date: 8 July 1999 (08.07.99)</p> <p>(30) Data relating to the priority: 198 31 869.3 16 July 1998 (16.07.98) DE</p> <p>(71) Applicant (for all designated States except US): MERCK PATENT GMBH [DE/DE]; Frankfurter Strasse 250, D-64293 Darmstadt (DE).</p> <p>(72) Inventors; and</p> <p>(75) Inventors/Applicants (US only): SCHWEINFURTH, Ralf [DE/DE]; Barlachstrasse 27, D-69168 Wiesbach (DE). MAURER, Uta [DE/DE]; Liebigstrasse 26, D-64293 Darmstadt (DE).</p> <p>(74) Joint Representative: MERCK PATENT GMBH; Postfach, D-64271 Darmstadt (DE).</p>		<p>(81) Designated states: AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ZA, ZW, ARIPO Patent (GH, GM, KE, LS, MW, SD, SL, SZ, UG, ZW), Eurasian Patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European Patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI Patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN TD, TG).</p> <p><b>Published</b></p> <p>With the International Search Report.</p> <p>Before expiry of the period provided for amending the claims, will be republished if such amendments are received.</p>

As printed

(54) Title: COLORATION WITH PEARLY LUSTER PIGMENTS IN THE FOOD AND PHARMACEUTICAL SECTORS

(54) Bezeichnung: FARBGEBUNG MIT PERLGLANZPIGMENTEN IM LEBENSMITTEL- UND PHARMABEREICH

(57) Abstract

The invention relates to the use of titanium oxide and/or iron oxide pigments on the basis of lamella-shaped substrates for coloring food and pharmaceutical products.

(57) Zusammenfassung

Die vorliegende Erfindung betrifft die Verwendung von Titandioxid- und/oder Eisenoxid-Pigmenten auf Basis plättchenförmiger Substrate zur Einfärbung von Lebensmittel- und Pharmaerzeugnissen.

Colouring using pearlescent pigments in the food and  
pharmaceutical sectors

5 The present invention relates to the use of 0.005% to 15%  
by weight of titanium dioxide pigments and/or iron oxide  
pigments based on platy substrates for colouring food  
products or pharmaceutical products characterized in that  
the platy substrate is a mica platelet,  $\text{Al}_2\text{O}_3$  platelet,  
 $\text{TiO}_2$  platelet or  $\text{SiO}_2$  platelet.

10 In addition to their functional applications,  
pearlescent pigments and interference pigments are also  
increasingly being used for the visual enhancement of  
products, for example in cosmetics, since fine colours  
15 and effects give rise to pleasing subjective  
impressions on the part of the viewer and consumer.  
Since, in the production of pearlescent pigments, for  
example for decorative cosmetics, the strictest  
requirements are made as to the purity and quality of  
20 the pigments, pearlescent pigments should also be  
usable in the food sector for improving the colouring  
effect or for colouring.

25 It would thus be desirable to expand the palette of the  
previously known colours in the colouring of food  
products and pharmaceutical products using pearlescent  
pigments or interference pigments, as the result of which  
the products experience an additional sensorially  
perceptible enhancement. The visual enhancement is  
30 valuable in particular for pharmaceutical products, since  
a clearer differentiation is made possible between  
differently coloured tablets, dragees, etc.

35 Surprisingly, it has now been found that for the visual  
enhancement of food products and pharmaceutical  
products, platy substrates coated with titanium dioxide  
and/or iron oxide are outstandingly suitable. Iron  
oxide pigment is taken to mean platy substrates coated  
not only with  $\text{Fe}_2\text{O}_3$  but also with  $\text{Fe}_3\text{O}_4$ . In particular,  
the combination of  $\text{TiO}_2$  pigments and/or  $\text{Fe}_3\text{O}_4$  pigments

with natural or nature-identical dyes, colour pigments or colorant fruit extracts and plant extracts give the food product an interesting new colour. By means of the colouring of food products, simultaneously desires for novel variants and nuances in colours as the result of novel trends in fashion can be taken into account.

The invention thus relates to the use of 0.005% to 15% by weight of titanium dioxide pigments and/or iron oxide pigments based on platy substrates for colouring food products or pharmaceutical products characterized in that the platy substrate is a mica platelet,  $\text{Al}_2\text{O}_3$  platelet,  $\text{TiO}_2$  platelet or  $\text{SiO}_2$  platelet.

The invention further relates to process for the production of food products and pharmaceutical products coloured by 0.005 to 15% by weight of titanium dioxide pigments and/or iron oxide pigments based on platy substrates, characterized in that the titanium oxide pigment and/or iron oxide pigment is a mica platelet,  $\text{Al}_2\text{O}_3$  platelet,  $\text{TiO}_2$  platelet or  $\text{SiO}_2$  platelet which is added to the product to be coloured alone or in combination with other pigments or colorants in the desired quantitative ratios, simultaneously or successively, during or after their production.

The invention also relates to food products and pharmaceutical products comprising 0.005 to 15% by weight titanium dioxide pigments and/or iron oxide pigments based on platy substrates as colorant, characterized in that the platy substrate is a mica platelet,  $\text{Al}_2\text{O}_3$  platelet,  $\text{TiO}_2$  platelet or  $\text{SiO}_2$  platelet.

The coloured foods and pharmaceutical products are distinguished by a novel colouring effect which is based on light refraction by the pearlescent pigments and causes pleasant subjective impressions in the viewer and consumer. This optical effect is not possible using the colorants which are currently

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permitted in the food sector. In contrast to colour pigments which are permitted in the food sector, for example vegetable carbon E153, the titanium dioxide pigments and iron oxide pigments based on platy substrates may be dispersed very readily into the medium to be pigmented. Furthermore, the products thus coloured are distinguished by an increased protection from light and moisture. Vitamin preparations in particular have a longer shelf life. In the case of the colouring of tablets, in many cases a delayed release of active compounds has been observed.

It has been found that even at very low amounts of titanium oxide pigments and/or iron oxide pigments novel interesting colours and simultaneously novel properties can be imparted to the foods or pharmaceutical products. Outstanding results are achieved even when the product is coloured with 0.005 to 15.0% by weight, preferably 0.01 to 6.0% by weight,

In the event that the pigment is added directly to the food product or pharmaceutical product during manufacture, the amount of pigment used is preferably 0.005 to 4% by weight. In the case of the surface treatment of foods or tablets, the application range is 0.02 to 15.0% by weight, preferably 0.5 to 6.0% by weight, based on the colorant solution or coating solution.

10

The black iron oxide pigments used are magnetite-coated natural or synthetic mica platelets, talc, kaolin,  $\text{SiO}_2$  platelets or  $\text{TiO}_2$  platelets. Particularly preferably, finely divided iron oxide pigments are used to colour the products, preferably having a particle size in the range from 0.01 to 200  $\mu\text{m}$ , in particular from 0.1 to 100  $\mu\text{m}$ . Black pearlescent pigments of this type are disclosed, for example, by the patents and patent applications P 23 13 331, P 36 17 430 and JP 90-246314. These pigments are commercially available, for example, under the trademark Candurin® Black Fine from Merck KGaA, Darmstadt.

$\text{TiO}_2$  pigments and  $\text{Fe}_2\text{O}_3$  pigments based on platy substrates, for example natural and synthetic mica, glass platelets,  $\text{TiO}_2$  platelets,  $\text{SiO}_2$  platelets and  $\text{Al}_2\text{O}_3$  platelets are disclosed, for example, by German patents and patent applications DE 14 67 468, DE 19 59 998, DE 20 09 566, DE 22 14 545, DE 22 15 191, DE 22 44 298, DE 23 13 331, DE 25 22 572, DE 31 37 808, DE 31 37 809, DE 31 51 343, DE 31 51 354, DE 31 51 355, DE 32 11 602, DE 32 53 017, WO 93/08237, DE 196 18 564 and EP 0 763 573. The substrates in these patents are coated with one, two, three, four, five or more metal oxide layers. In particular, preference is given to pigments which comprise one or two layers of  $\text{TiO}_2$  and/or  $\text{Fe}_2\text{O}_3$ . The particle sizes of the pigments are preferably  $\leq 200 \mu\text{m}$ , in particular  $\leq 100 \mu\text{m}$ .

By adding pearlescent pigments and interference pigments which are permitted for the food sector, and/or natural/nature-identical dyes, colour pigments or colorant fruit extracts and plant extracts, the  
5 colouring effect of the titanium dioxide pigments and/or iron oxide pigments can be intensified in the product and simultaneously novel colouring effects can be achieved.

10 In particular, the colouring of food products with a pigment mixture consisting of  $\text{Fe}_3\text{O}_4$  pigments with pearlescent pigments and/or interference pigments based on mica platelets,  $\text{Al}_2\text{O}_3$  platelets,  $\text{SiO}_2$  platelets or  
15  $\text{TiO}_2$  platelets which are coated with  $\text{TiO}_2$  and/or  $\text{Fe}_2\text{O}_3$ , imparts interesting colouring effects to the products. Very particular preference is given to pigment mixtures comprising an  $\text{Fe}_3\text{O}_4$  pigment based on mica.

Combining  $\text{Fe}_3\text{O}_4$  pigments with other pearlescent pigments  
20 such as gold pigments, silver pigments or interference pigments intensifies the respective colouring effect of the pigments. This synergy extends considerably the colouring possibilities of the product to be pigmented, without other natural or nature-identical dyes having  
25 to be additionally used.

The pearlescent pigments and interference pigments permitted for the food sector are commercially available, for example, under the trademark Candurin®  
30 from Merck KGaA.

The total concentration of all pigments in the product to be pigmented should also not exceed 12% by weight, based on the product. The concentration is generally  
35 dependent on the specific application. The mixing ratio of the  $\text{TiO}_2$  pigments or  $\text{Fe}_3\text{O}_4$  pigments with a further pigment component depends on the desired effect and is generally 20:1 to 1:20, preferably 5:1, in particular 1:1. The pigment component can be one or more

pearlescent pigments or interference pigments. In particular preference is given to  $\text{TiO}_2$ -mica pigments,  $\text{Fe}_2\text{O}_3$ -mica pigments and  $\text{TiO}_2$ - and  $\text{Fe}_2\text{O}_3$ -coated mica pigments. In addition, preference is given to  $\text{TiO}_2$ -  
5 and/or  $\text{Fe}_2\text{O}_3$ -coated or uncoated  $\text{SiO}_2$  platelets or  $\text{TiO}_2$  platelets.

The colouring effect in food products and pharmaceutical products is improved in particular when  
10 black iron oxide pigments are combined with gold pigments, silver pigments and interference pigments based on  $\text{TiO}_2$ - or  $\text{TiO}_2/\text{Fe}_2\text{O}_3$ -coated mica pigments. The content of  $\text{Fe}_3\text{O}_4$  pigments in the product should, in this preferred embodiment, preferably be 0.005-2% by weight.

15 In addition to the pearlescent pigments and/or interference pigments, all natural or nature-identical dyes known to those skilled in the art can be added as further colouring component to the titanium dioxide  
20 pigments and/or iron oxide pigments. In particular those which may be mentioned here are: E 101, E 104, E 110, E 124, E 131, E 132, E 140, E 141, E 151, E 160a.

25 In addition, other colouring pigments can be added to the platy pearlescent pigments, for example E 171, E 172, E 153.

The content of dyes based on the product is in the  
30 range from 0.5 to 25% by weight. Also, fruit extracts and plant extracts can be used as dye, for example carrot juice, beetroot juice, elderberry juice, hibiscus juice, paprika extract, aronia extract.

35 The pharmaceutical products and food products are coloured by adding the titanium dioxide pigment and/or iron oxide pigment to the product to be coloured alone or in combination with other pigments or colorants in the desired quantitative ratios, simultaneously or



successively, during or after their production. Laborious grinding and dispersion of the pigments is not necessary.

5 Products suitable for colouring which may be mentioned  
are in particular coatings on all types of foods, in  
particular pigmented sugar coatings and shellac  
coatings (alcoholic and aqueous), coatings containing  
oils and waxes, containing gum arabic and cellulose  
10 types (e.g. HPMC = hydroxypropyl methyl cellulose),  
incorporation into or application onto confectionery,  
cake decorations, compressed tablets, pan-coated  
products, chewing gums, gums, fondant products,  
marzipan products, fillings, cocoa icings and fat  
15 icings, chocolate and chocolate-containing products,  
ice cream, cereals, snack products, coating  
compositions, gateaux presentation plates [sic],  
hundreds and thousands, sugar crystals, jelly and  
gelatin products, sweets, liquorice, icing, candyfloss,  
20 fat, sugar and baker's cream compositions, puddings,  
desserts, flan glazing, cold sweet soups, beverages  
containing stabilizing additives such as carboxy methyl  
cellulose, acidified and non-acidified milk products  
such as quark, yoghurt, cheese, cheese rinds, sausage  
25 casings, etc.

A further large field of use is the pharmaceutical and  
OTC sector for colouring tablets, gelatin capsules,  
sugar-coated tablets, ointments, cough syrup, etc. In  
30 combination with customary coatings such as  
polymethacrylates and celluloses, for example HPMC, the  
pigments can be used in many ways for colouring.

In the case of pan-coated or otherwise coated food  
35 products and pharmaceutical products, it is possible to  
combine the titanium dioxide pigments and iron oxide  
pigments with flavourings (powdered flavourings or  
liquid flavourings) and/or with sweeteners, for example

aspartame, in order to accentuate the visual effect also in terms of flavour.

5 The invention thus relates to all formulations from the food sector and pharmaceutical sector comprising the titanium dioxide pigment and/or iron oxide pigment alone or in combination with other pigments/pigment mixtures or dyes (natural or nature-identical) as colorants.

10 The examples below are intended to describe the invention without restricting it, however.

### Examples

15

#### Example 1 - Hard caramel manufacturing details

##### 1. Example formula containing Isomalt

20			Obtainable from:
	Isomalt Merck	75%	Palatinit GmbH,
	Patent GmbH		Mannheim
	Water	24.005%	
	Aspartame	0.0075%	Worlée, Hamburg
25	Acesulfame-K	0.0075%	Nutrinova,
			Frankfurt a.M.
	Candurin® Silver	0.144% (0.2% based	Merck KGaA,
	Sheen <sup>1</sup>	on the	Darmstadt
		pouring mass)	
30	Candurin® Black	0.036% (0.05% based	Merck KGaA,
	Fine <sup>2</sup>	on the	Darmstadt
		pouring mass)	
	Citric acid	0.5%	Merck KGaA,
			Darmstadt
35	Flavouring	0.3% (peach 9/030307)	Dragoco,
			Holzminden

<sup>1</sup> (TiO<sub>2</sub>-mica pigment of particle size 5-25 µm)

<sup>2</sup> (Fe<sub>3</sub>O<sub>4</sub>-mica pigment of particle size < 15 µm)

Water is heated with Isomalt M to 165°C and the solution is then allowed to cool to 145°C. After addition of citric acid, aspartame, Acesulfame-K, the pigments Candurin® Black Fine and Candurin® Silver Sheen and the flavouring are stirred in. Finally the hot mixture is poured into greased moulds using a pouring funnel. The hard caramels are allowed to cool for 1 hour.

10 Further embodiments:

- It is frequently advisable to dissolve the ingredients in advance using some of the water in order to avoid lump formation.
- The pigments Candurin® Silver Sheen and Candurin® Black Fine can also be heated directly with the water and Isomalt. No loss of colour occurs.

## 2. Example formula containing Isomalt

20	Isomalt Merck	75%	Obtainable from:
	Patent GmbH		Palatinit GmbH,
	Water	24.041%	Mannheim
	Aspartame	0.0075%	Worlée, Hamburg
25	Acesulfame-K	0.0075%	Nutrinova,
			Frankfurt a.M.
	Candurin®	0.144% (0.2% based	Merck KGaA,
	Honeygold <sup>3</sup>	on the	Darmstadt
		pouring mass)	
30	Citric acid	0.5%	Merck KGaA,
			Darmstadt
	Flavouring	0.3% (peach 9/030307)	Dragoco,
			Holzminden

<sup>3</sup> (TiO<sub>2</sub>/Fe<sub>2</sub>O<sub>3</sub>-mica pigment of particle size 5-25 µm)

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Water is heated with Isomalt M to 165°C and the solution is then allowed to cool to 145°C. After addition of citric acid, aspartame, Acesulfame-K, Candurin® Honeygold and the flavouring are stirred in.

Finally, the hot mass is poured into greased moulds using a pouring funnel. The hard caramels are allowed to cool for 1 hour.

5 Further embodiments:

- Frequently it is advisable to dissolve the ingredients in advance using some of the water in order to prevent lump formation.
- The pigment Candurin® Honeygold can also be heated directly with the water and Isomalt.

3. Example formula containing sugar

			Obtainable from:
15	Sugar	41%	Südzucker
	Water	17.077%	
	Glucose syrup	41% C* Sweet	Cerestar, Krefeld
	Candurin® Silver	0.082% (0.1% based	Merck KGaA,
	Sheen <sup>1</sup>	on the	Darmstadt
20		pouring mass)	
	Candurin® Black	0.041% (0.05% based	Merck KGaA,
	Fine <sup>2</sup>	on the	Darmstadt
		pouring mass)	
	E 104 1:100 dilution	0.4% Sikovit	BASF,
25			Ludwigshafen
	Flavouring	0.4% (banana 9/030388)	Dragaco [sic],
			Holzminden

<sup>1</sup> (TiO<sub>2</sub>-mica pigment of particle size 5-25 µm)

<sup>2</sup> (Fe<sub>3</sub>O<sub>4</sub>-mica pigment of particle size < 15 µm)

30

The sugar is heated with the water to 100°C and then glucose syrup is added. The solution is then heated to 145°C. After addition of Candurin® Silver Sheen, Candurin® Black Fine, dye solution and the flavouring,

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the caramel solution is poured into greased moulds using a pouring funnel. Finally, the mixture is allowed to cool for two hours. The Candurin® pigments can be either mixed with the sugar or added in a mixture with

the glucose syrup. This variant contains no acid, since this would make the caramelization too intense.

#### 4. Example formula containing sugar

5			Obtainable from:
	Sugar	41%	Südzucker
	Water	17.118%	
	Glucose syrup	41% C <sup>*</sup> Sweet	Cerestar, Krefeld
10	Candurin <sup>®</sup> Wine Red <sup>4</sup>	0.082% (0.1% based on the pouring mass)	Merck KGaA, Darmstadt
	E 104 1:100 dilution	0.4% Sikovit	BASF, Ludwigshafen
15	Flavouring	0.4% (banana 9/030388)	Dragaco [sic], Holzminden
	<sup>4</sup> (Fe <sub>2</sub> O <sub>3</sub> -mica pigment of particle size 10-60 µm)		

20 The sugar is heated with the water to 100°C and then glucose syrup is added. The solution is then heated to 145°C. After addition of Candurin<sup>®</sup> Wine Red, dye solution and the flavouring, the caramel solution is poured into greased moulds using a pouring funnel. Finally, the mixture is allowed to cool for two hours.

25 The Candurin<sup>®</sup> pigment can be either mixed with the sugar or added as a mixture with the glucose syrup. This variant contains no acid, since this would make the caramelization too intense.

#### 30 Example 2 - Manufacture of gelatin articles

##### 1. Example formula

			Obtainable from:
35	Water	10.48%	
	Sugar	31.45%	Südzucker
	Glucose syrup	31.45% C <sup>*</sup> Sweet	Cerestar, Krefeld
	Candurin <sup>®</sup> Wine Red	0.38% (0.4% based on the	Merck KGaA, Darmstadt

		pouring mass)	
	Citric acid 1:1	2.51%	Merck KGaA,
	dilution		Darmstadt
	Gelatin	7.86% 260 Bloom	DGF, Eberbach
5	Water	15.748%	
	Flavouring	0.122% (blackcurrant	Dragoco,
		9/695750)	Holzminden

10 The gelatin is first softened with twice the amount of water at 60°C. Sugar and water are heated to 100°C, then the glucose syrup is added. The mixture is heated further to 120°C and is then allowed to cool to approximately 85°C. The Candurin® pigment, the citric acid, the flavouring and the gelatin solution are

15 stirred in, and the deaerated gelatin mixture is charged into greased moulds using the pouring funnel. The product is allowed to cool for approximately 16 hours.

20 Further embodiments:

- The Candurin® pigment can here again be already mixed directly with the sugar or introduced together with the glucose syrup.
  - Instead of pouring into moulds, the traditional
- 25 method using negative moulds in moulding powder can be used in this case to produce gelatin articles.

## 2. Example formula

30			Obtainable from:
	Water	10.508%	
	Sugar	31.45%	Südzucker
	Glucose syrup	31.45% C* Sweet	Cerestar, Krefeld
	Candurin® Blueberry	0.38% (0.4% based	Merck KGaA,
35	Sugar <sup>5</sup>	on the	Darmstadt
		pouring mass)	
	E 153 (vegetable	0.038% (0.04% based	Dr. Marcus
	carbon/Carbon	on the	
	medicinales)	pouring mass)	

	Citric acid 1:1	2.51%	Merck KGaA, Darmstadt
	dilution		
	Gelatin	7.86% 260 Bloom	DGF, Eberbach
	Water	15.682%	
5	Flavouring	0.122% (blackcurrant 9/695750)	Dragoco, Holzminden

\*5 (TiO<sub>2</sub>-mica pigment of particle size 10-60 µm)

10 The gelatin is first softened with the same amount of water at 60°C. Sugar and water are heated to 100°C, then the glucose syrup is added. The mixture is heated further to 120°C and is then allowed to cool to approximately 85°C. The Candurin® pigment, the citric acid, the flavouring and the gelatin solution are  
15 stirred in, and the deaerated gelatin mixture is charged into greased moulds using the pouring funnel. The product is allowed to cool for approximately 16 hours.

20 Further embodiments:

- The Candurin® pigment can here again be already mixed directly with the sugar or introduced together with the glucose syrup.
- Instead of pouring into moulds, the traditional  
25 method using negative moulds in moulding powder can be used in this case to produce gelatin articles.

### Example 3 - Dragees

30 a) Cores to be coloured: white dragee cores (liquorice rods with a hard sugar coating)  
Solution for coating the dragees:

			Obtainable from:
35	- alcoholic shellac solution	95.62%	Wolff & Olsen
	- Candurin® Silver Lustre* <sup>1</sup>	4.00%	Merck KGaA, Darmstadt
	- Candurin® Black Fine* <sup>2</sup>	0.38%	Merck KGaA, Darmstadt

\*<sup>1</sup> (TiO<sub>2</sub>-mica pigment of particle size 10-60 µm)

\*<sup>2</sup> (Fe<sub>3</sub>O<sub>4</sub>-mica pigment of particle size < 15 µm)

The Candurin® pigments are evenly distributed in the shellac and sprayed onto the dragees which are slowly rotating in the dragee pan. The dragees are continuously dried using cold air. The spraying is continued until the desired colour coverage is achieved. Finally, the cores are taken out of the pan and dried on racks for approximately 12 hours.

- 10 b) Cores to be coloured: black dragee cores (chewing gum balls)

Solution for coating the dragees:

		Obtainable from:
15	- alcoholic shellac solution 95%	Kaul GmbH
	- Candurin® Caramel* 5%	Merck KGaA, Darmstadt
	(Fe <sub>2</sub> O <sub>3</sub> -mica pigments of particle size 10-60 µm)	

20 The Candurin® pigment is evenly distributed in the shellac and sprayed onto the dragees which are slowly rotating in the dragee pan. Spraying is continued until the desired colour coverage is achieved. Drying is performed continuously using cold air in order to prevent the cores from sticking together. Finally, the  
25 cores are taken out of the pan and dried on racks for approximately 12 hours.

- c) - Coating liquorice nibs with a white hard sugar shell

30

Cores to be coloured: liquorice nibs having a white hard sugar shell

Solution for coating the dragees:

35		Obtainable from:
	- alcoholic shellac solution 95.75%	
		Capol 425 Kaul GmbH
	- Candurin® Buttergold* <sup>1</sup> 4%	Merck KGaA, Darmstadt
	- Candurin® Black Fine 0.25%	Merck KGaA, Darmstadt



(TiO<sub>2</sub>/Fe<sub>2</sub>O<sub>3</sub>-mica pigment of particle size 10-60 µm)

The cores are coated in a similar manner to Example 3 a).

5

d) - Pan-coating of Viennese nuts (white, hard sugar shell with hazelnut core)

Solution for coating the dragees:

10

		Obtainable from:
- alcoholic shellac solution	96.0%	Wolff & Olsen
- Candurin® Silver Lustre*	4%	Merck KGaA, Darmstadt
* (TiO <sub>2</sub> -mica pigment of particle size 10-60 µm)		

15 The Candurin® pigment is distributed evenly in the shellac solution. It is applied in a similar manner to Example 3 a).

e) - Starting material: white chocolate drops

20

An aqueous Candurin®/HPMC solution is used for the spray application.

25 - The Candurin® pigments are stirred into water. Then, if appropriate, additional dyes, flavourings or sweeteners are then added. Finally, the film-forming agent (HPMC) is added to the suspension. As a result of the increasing viscosity, the stirrer speed must also be correspondingly increased. After approximately 40-60 minutes, the HPMC is completely dissolved and the solution can then be sprayed onto the dragees.

30

- The spray application can be performed in the pan-coating drum or in customary coating systems, with care being taken to ensure an appropriately matched continuous drying air stream (temperatures/volumes).

35

- As soon as the desired colouring effect is achieved, the dragees are discharged. They are then dried on racks.

Spray solution:

Sepifilm Lp10	6.0%	Seppic
Candurin® paprika	5.0%	Merck KGaA, Darmstadt
Flavouring	0.5% (vanilla 9/024233)	Dragoco, Holzminden
Water	88.5%	

(Fe<sub>2</sub>O<sub>3</sub>-mica pigment of particle size 10-60 µm)

- 5 f) Starting material: white sugar-pan-coated almonds

The spray solution is prepared and applied as in e)

Spray solution:

Sepifilm Lp10	5.0%	Seppic
Candurin® Silver Lustre	4.0%	Merck KGaA, Darmstadt
Sepisperse M5062	1.0%	Seppic
Water	90.0%	

- 10 (TiO<sub>2</sub>-mica pigment of particle size 10-60 µm)

- g) Starting material: white sugar-pan-coated almonds

The spray solution is prepared and applied as in e)

15

Spray solution:

Sepifilm Lp10	6.0%	Seppic
Candurin® Silver Lustre	4.0%	Merck KGaA, Darmstadt
E153	0.2%	Dr. Marcus
Water	89.8%	

(TiO<sub>2</sub>-mica pigment of particle size 10-60 µm)

- h) Starting material: red sugar-pan-coated almonds

20

An aqueous Candurin®/gum arabic solution is used for the spray application.

The Candurin® pigments are stirred into the gum arabic

- 25 solution and are then sprayed onto the dragees rotating

in the pan-coating drum. Drying should be performed continuously here using cold air. When the desired colouring effect is achieved, the spraying operation is terminated. The coloured dragees can then further be coated with a shellac film to avoid sticking together.

Spray solution:

Gum arabic solution	82.0%	Kaul
Candurin® Wine Red*	8.0%	Merck KGaA, Darmstadt
Water	10.0%	

\* (Fe<sub>2</sub>O<sub>3</sub>-mica pigment of particle size 10-60 µm)

10 Example 4 - Jelly dessert

		Obtainable from:
	Calcium lactate	0.05% Merck KGaA
	Genugel LC4N	0.6% Hercules
15	Tetrasodium phosphate	0.15% Merck KGaA
	Citric acid crystalline	0.38% Merck KGaA
	gellan	0.06% Kelco
	Tripotassium phosphate	0.05% Merck KGaA
	Sucrose	15% Südzucker
20	Candurin® Silver Sparkle <sup>1</sup>	0.025% Merck KGaA
	Flavouring and colour optional	
	Water	83.685%

<sup>1</sup> (TiO<sub>2</sub>-mica pigment of particle size 20-150 µm)

25 All components are heated to 95°C and kept at this temperature for 3-5 minutes with stirring. They are then allowed to cool with gentle stirring to 40-45°C. The jelly dessert is then poured into moulds and if necessary it is cooled.

Pharmaceutical applications:

Example 5

- 5 a) Initial weight 1 kg of white tablets  $d = 8$  mm,  
G = 200 mg

Solution for film coating:

6%	Sepifilm Lp10 (Mixture of hydroxypropyl methyl cellulose, stearic acid and microcrystalline cellulose)	Seppic
5%	Candurin® Caramel	Merck KGaA, Darmstadt
89%	Water	

\* ( $\text{Fe}_2\text{O}_3$ -mica pigment of particle size 10-60  $\mu\text{m}$ )

10

Total amount applied: 200 g

This corresponds to 1.2 mg of polymer/ $\text{cm}^2$  of tablet  
surface area

- 15 b) Initial weight 1 kg of white tablets  $d = 8$  mm,  
G = 200 mg

Solution for film coating (100 g):

6%	Sepifilm Lp10	Seppic
4%	Candurin® Silver Lustre	Merck KGaA
1%	Sepisperse M5062 (Dye paste: $\text{TiO}_2$ , natural or nature- identical dyes, stabilizer)	Seppic
89%	Water	

\* ( $\text{TiO}_2$ -mica pigment of particle size 10-60  $\mu\text{m}$ )

20

Total amount applied: 200 g

This corresponds to 1.2 mg of polymer/ $\text{cm}^2$  of tablet  
surface area

- 25 Production of the film-coating solution:

- The Candurin® pigments are stirred into water. If appropriate, additional dyes are then added. Finally, the film-forming agent (HPMC) is dispersed in the suspension. The increasing viscosity necessitates the stirrer viscosity also being correspondingly increased. After approximately 40-60 minutes, the HPMC is completely dissolved and the solution can then be sprayed onto the tablets.
- The spray application is made using a customary standard coating process.

### Sugar confectionery

#### Example 6:

##### a) Liquorice products:

Raw material: extruded liquorice products

- The oiled liquorice products are sprayed with a Candurin®/shellac solution in a pan-coating drum. At the same time they are dried using cold air. As soon as the desired colouring effect is achieved, application is halted and the coloured liquorice products are discharged from the pan.

#### Spray solution:

Alcoholic shellac solution	97%	Kaul
Candurin® Kiwi Sugar*	3%	Merck KGaA, Darmstadt

\* (TiO<sub>2</sub>-mica pigment of particle size 10-60 µm)

##### b) Menthol pastilles

Raw material: menthol pastilles

- A Candurin®/shellac solution (aqueous) is sprayed onto the rotating pastilles in the pan-coating drum. Drying is performed continuously with warm air here. As soon as the desired colouring effect is achieved,

application is terminated and the coloured pastilles are discharged from the pan.

Spray solution:

Alcoholic shellac solution	96%	Warner Jenkinson
Candurin® Silver Lustre*	4%	Merck KGaA, Darmstadt

5 \* (TiO<sub>2</sub>-mica pigment of particle size 10-60 µm)

c) Marzipan roses (cake decoration)

Raw material: red marzipan roses

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The cake decorations are sprayed with a Candurin®/shellac solution until the desired colour application is achieved. Subsequent drying with cold air is possible.

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Spray solution:

Alcoholic shellac solution	97%	Wolff & Olsen
Candurin® Blueberry Sugar*	3%	Merck KGaA, Darmstadt

\* (TiO<sub>2</sub>-mica pigment of particle size 10-60 µm)

d) Sherbet sweets

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Raw material: sherbet sweets, white

The sherbet sweets are sprayed with a Candurin®/shellac solution until the desired colour application is achieved. Subsequent drying with cold air is possible.

25

Spray solution:

Alcoholic shellac solution	94%	Kaul
Candurin® Silver Sparkle*	6%	Merck KGaA, Darmstadt

\* (TiO<sub>2</sub>-mica pigment of particle size 20-150 µm)

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The reference to any prior art in this specification is not, and should not be taken as, an acknowledgement or any form of suggestion that the prior art forms part of the common general knowledge in Australia.

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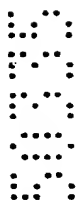
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The claims defining the invention are as follows:

1. Use of 0.005% to 15% by weight of titanium dioxide pigments and/or iron oxide pigments based on transparent platy substrates for colouring food products or pharmaceutical products characterized in that the platy substrate is a mica platelet,  $\text{Al}_2\text{O}_3$  platelet, or  $\text{SiO}_2$  platelet.
2. Use of titanium dioxide pigments and/or iron oxide pigments according to Claim 1 characterized in that the iron oxide pigment is a platy substrate coated with  $\text{Fe}_2\text{O}_3$  or  $\text{Fe}_3\text{O}_4$ .
3. Use of titanium dioxide pigments and/or iron oxide pigments according to Claim 1 or Claim 2, characterized in that the titanium dioxide pigment and the iron oxide pigment can be mixed in a ratio of 1:20 to 20:1.
4. Use of titanium dioxide pigments and/or iron oxide pigments according to anyone of Claims 1 to 3, characterized in that titanium dioxide pigments and/or iron oxide pigments are combined with one or more pearlescent pigments,  $\text{SiO}_2$  platelets, natural and/or nature-identical colorants, colour pigments, dyes, flavourings, sweeteners or natural plant or fruit extracts.
5. Use of titanium dioxide pigments and/or iron oxide pigments according to Claim 4, characterized in that the titanium dioxide pigments and/or iron oxide pigments are used in combination with one or more pearlescent pigments based on mica coated with  $\text{TiO}_2$ ,  $\text{Fe}_2\text{O}_3$  or a  $\text{TiO}_2/\text{Fe}_2\text{O}_3$  mixture.



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6. Use of titanium dioxide pigments and/or iron oxide pigments according to Claim 4 or 5, characterized in that the iron oxide pigment is a mica platelet,  $\text{SiO}_2$  platelet or  $\text{TiO}_2$  platelet coated with  $\text{Fe}_3\text{O}_4$ .
- 5 7. Use of titanium dioxide pigments and/or iron oxide pigments according to anyone of Claims 1 to 6, characterized in that the food product or pharmaceutical product is furnished with a coating of shellac, oils, waxes, gum arabic, celluloses, 10 polymethacrylates or icing comprising titanium dioxide pigments and/or iron oxide pigments and if appropriate other pigments and/or colorants.
- 15 8. Use of titanium dioxide pigments and/or iron oxide pigments according to any one of Claims 1 to 7, for colouring tablets, gelatine capsules, sugar-coated tablets, ointments or cough syrups.
- 20 9. Process for the production of food products and pharmaceutical products coloured by 0.005 to 15% by weight of titanium dioxide pigments and/or iron oxide pigments based on transparent platy 25 substrates, characterized in that the titanium oxide pigment and/or iron oxide pigment is a mica platelet,  $\text{Al}_2\text{O}_3$  platelet, or  $\text{SiO}_2$  platelet which is added to the product to be coloured alone or in combination with other pigments, coatings and/or colorants in the desired quantitative ratios, 30 simultaneously or successively, during or after their production.
10. Food products and pharmaceutical products comprising 0.005 to 15% by weight titanium dioxide pigments and/or 35 iron oxide pigments based on transparent platy substrates as colorant, characterized in that the transparent platy

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substrate is a mica platelet,  $\text{Al}_2\text{O}_3$  platelet, or  $\text{SiO}_2$  platelet.

11. Uses of 0.005 to 15% by weight of titanium dioxide pigments and/or iron oxide pigments based on transparent platy substrates, substantially as hereinbefore described with reference to the Examples.

12. Processes for the production of food products and pharmaceutical products, substantially as hereinbefore described with reference to the Examples.

DATED this 17th day of July, 2003

MERCK PATENT GMBH  
By its Patent Attorneys  
DAVIES COLLISON CAVE

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